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Morten W. Ryberg*, Mikołaj Owsianiak, Alexis Laurent, Michael Z. Hauschild

12 **Power generation from chemically cleaned coals: do environmental benefits of firing cleaner**
13 **coal outweigh environmental burden of cleaning?**

14

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24

25 **Abstract**

26 Power generation from high-ash coals is a niche technology for power generation, but coal cleaning
27 is deemed necessary to avoid problems associated with low combustion efficiencies and to
28 minimize environmental burdens associated with emissions of pollutants originating from ash.
29 Here, chemical beneficiation of coals using acid and alkali-acid leaching procedures is evaluated as
30 a potential coal cleaning technology employing life cycle assessment (LCA). Taking into account
31 the environmental benefits from firing cleaner coal in pulverized coal power plants and the
32 environmental burden of the cleaning itself, it is demonstrated that for a wide range of cleaning
33 procedures and types of coal, chemical cleaning generally performs worse than combustion of the
34 raw coals and physical cleaning using dense medium separation. These findings apply for many
35 relevant impact categories, including climate change. Chemical cleaning can be optimized with
36 regard to electricity, heat and methanol use for the hydrothermal washing step, and could have
37 environmental impact comparable to that of physical cleaning if the overall resource intensiveness
38 of chemical cleaning is reduced by a factor 5 to 10, depending on the impact category. The largest
39 potential of the technology is observed for high-ash lignites, with initial ash content above 30%, for
40 which the environmental benefits from firing cleaner coal can outweigh the environmental burden
41 of cleaning for some impact categories. We recommend for policy makers to use physical cleaning,
42 as this clearly perform environmentally better, but encourage further research into the chemical
43 cleaning process and optimization of the process as chemical cleaning may be necessary to comply

44 with current and emerging legislation on ash and sulphur content in coal where the removal
45 efficiency from physical cleaning is insufficient.

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47

48 **Broader context**

49 The use of fossil coal for generation of electricity is a major cause of many environmental problems
50 globally. Pulverized coal-fired power plants currently account for 97% of total coal-based
51 electricity generation globally, and are expected to constitute a significant proportion of the
52 environmental burden attributable to power generation also in the future. Chemical cleaning of
53 high-ash coals can potentially mitigate some environmental impacts from firing coal in pulverized
54 power plants, as it allows reaching higher ash removal efficiencies as compared to physical
55 cleaning, but virtually nothing is known about whether environmental benefits from firing cleaner
56 coal outweigh environmental burdens of cleaning. Here, life cycle assessment (LCA) is employed
57 to evaluate chemical beneficiation of coals using acid and alkali-acid leaching procedures. The
58 results showed that demineralization is generally not a feasible option for mitigating environmental
59 impacts, including impacts from climate change, except in few cases where regional and local
60 impacts were improved for high-ash coal systems. This work highlights the current issues
61 associated with chemical cleaning technologies and provides recommendations for stakeholders to
62 resolve these.

63

64 **1. Introduction**

65 The use of fossil coal for generation of electricity is a major cause of environmental problems
66 globally. Using life cycle impact assessment, Laurent and Espinosa¹ showed that, while coal
67 represented 41% of the global electricity produced in 2011, it contributed to more than 70% of the
68 associated environmental burden for nearly all impacts on human health and ecosystems. This
69 important contribution may decrease in the future, as the major focus of national policies is to
70 decrease the share of coal in the power mix and to improve environmental performance of coal-
71 based electricity generation^{2,3}. The latter include shutting down small, inefficient power plants,
72 imposing the deployment of cleaner coal technologies (CCTs) such as ultra-supercritical plants (*e.g.*
73 in India, China, or South Africa), enforcing more stringent emission control and standards with
74 respect to emissions of CO₂, SO₂, NO_x, particulate matter (PM), or mercury (*e.g.* in European

75 Union, U.S.A., or Indonesia), and introducing carbon capture and storage systems (*e.g.* in European
76 Union, U.S.A. and Canada) ^{4,5}.

77 Despite these initiatives, coal combustion in pulverized coal (PC) fired power plants, that
78 currently accounts for 97% of total coal-based electricity generation (of which ca. 76% is old,
79 subcritical power plants), can still be expected to take up a significant proportion of the total
80 environmental burden attributable to power generation ^{3,6}. Low-rank coals (with ash content above
81 30%) are relatively abundant as compared to high-rank coals and become increasingly important to
82 secure a stable coal supply for power generation in pulverized power plants. ^{7,8,9}. Their extraction
83 and firing becomes economically favorable as compared to import of high quality coals from
84 elsewhere ^{10,11}. Extraction and use of low-ranked coals thus takes place in Europe, U.S.A., China
85 and India today, mainly due to depletion of high quality coal reserves and extraction of low rank
86 coal being economically better than import of high quality coals. Power generation in some
87 important coal using countries, including India, China, Czech Republic, Poland, South Africa,
88 Romania and Turkey, currently relies strongly on these low-rank coals ¹²

89 Coal cleaning to reduce ash content, usually below 30%, is deemed capable to avoid problems
90 associated with low combustion efficiencies and minimize environmental burdens associated with
91 emissions of airborne pollutants ¹³. This is true for both old (subcritical) and newer (*e.g.*
92 supercritical or ultra-supercritical) combustion technologies. Coal cleaning is commercially done
93 using physical methods, such as grinding of the raw coal followed by gravity-based processes (*e.g.*
94 dense medium separation) and processes based on surface properties (*e.g.* froth flotation). These
95 methods have relatively low ash removal efficiencies and are generally not suitable to coals where
96 inherent ash-related minerals are finely disseminated in the coal matrix ^{7,13}. In contrast, chemical
97 cleaning using acid or alkali-acid leaching procedures are applicable to high-ash coals and allow
98 removing ash more efficiently than physical methods. Because of this, chemical coal cleaning is
99 seen as a complementary alternative to physical cleaning, allowing for achieving the required ash
100 removal rates that cannot be achieved by solely using physical cleaning. Since the 1980s, a total of
101 26 studies have been published with focus on technological aspects of chemical cleaning using acid
102 or alkali-acid leaching procedures (*e.g.* ^{14–18}; see Wijaya and Zhang¹⁹ and Meshram et al.¹³ for
103 reviews). These studies show a great potential for chemical cleaning of coals, with ash removal
104 efficiencies reaching up to ca. 97%, depending on the initial ash content and process conditions.
105 Chemical cleaning procedures are currently not widely employed at a commercial scale due to high
106 costs associated with the requirements of chemicals and the need for dewatering of the post-

107 demineralization fine-sized slurry. To date, only one pilot plant has been constructed in Australia,
108 which from 2010 to 2012 produced ultra clean coal (UCC, with ash content of ca. 0.1%) derived
109 from a bituminous coal for application in integrated gasification combined cycle ^{19,20}. In the future,
110 chemical cleaning procedures can however become attractive also for PC firing combustion
111 technologies, particularly for those coals for which physical beneficiation might not be a sufficient
112 method for ash removal.

113 Coal cleaning can both improve and decrease the overall environmental performance of coal
114 firing, depending on the type of environmental impact and stage of the coal combustion life cycle
115 considered. Table 1 shows the implications of coal cleaning using chemical methods on
116 environmental performance of power generation from cleaned coals. For example, reduced contents
117 of sulfur and ash in the coal due to chemical leaching will allow reducing emissions of SO₂ and PM,
118 thereby decreasing acidifying impacts on terrestrial ecosystems and damages to human health, and
119 increased combustion efficiencies are expected to reduce climate change impacts up to 5 % ²¹ due to
120 reduced emissions of CO₂ (per unit of electricity produced). By contrast, chemical leaching requires
121 provision of heat and electricity, which can increase climate change impacts and toxicity-related
122 impacts on human health and ecosystems due to the associated emissions of CO₂ and toxic
123 substances. These potential tradeoffs between the environmental benefits of firing cleaner coal and
124 the environmental burden of cleaning processes can be quantified using life cycle assessment
125 (LCA). In LCA, resource consumption and emissions of pollutants stemming from the extraction of
126 the raw materials, their manufacture and use or operations up to their end-of-life are inventoried and
127 translated into impact indicator scores using substance-specific characterization factors for various
128 life cycle impact categories. To date, LCA of power generation from coals cleaned chemically has
129 not been reported in the literature.

130 The aim of this study is to evaluate chemical cleaning of coals using acid and alkali-acid
131 leaching procedures as a potential coal cleaning technology. The focus is on chemical cleaning
132 because of the lack of knowledge about its environmental performance in coal-based power
133 generation; the goal is to provide a holistic environmental assessment of the technology to facilitate
134 well-informed decisions on use of the technology. To illustrate the potential of the technology, all
135 available variants of leaching procedures published so far for bituminous and sub-bituminous coals,
136 and lignites, were compared (i) to each other, (ii) to physical cleaning using dense medium
137 separation, and (iii) to conventional power generation from raw coals. The coals analysed in the

study vary with regard to the initial ash content (prior to leaching), and also include high-ash lignites with initial ash content above 30%.

Table 1. Changes introduced by switching from use of raw coal to coal cleaned chemically and their expected consequences for overall environmental performance.

Differences induced by switching from raw coal to coal cleaned chemically	Expected consequences on environmental performance
<i>Expected reduction in environmental impacts:</i>	
Reduced airborne emissions from the power plant	Part of the ash and sulfur embedded in coal is emitted from the power plant as particulate matter (PM) in fly ashes, and as SO _x . Lower ash and sulfur content will thus reduce PM and SO _x emissions ²² , which is expected to reduce acidification impact on terrestrial ecosystems and human health effects of particulate matter.
Coal content of hazardous metals like antimony, arsenic, cadmium, chromium, copper, cobalt, mercury, lead, manganese and nickel, generally present in trace amounts is reduced through the cleaning process ²³	The hazardous trace elements in the coal are emitted during the combustion or as part of the flue gas residue disposal. Removal of these pollutants will reduce toxic impact to humans and ecosystems. The environmental benefits of this removal cannot be included in this study because of missing data on the content of these elements in the uncleaned coal and their removal during coal cleaning.
Increased overall power plant efficiency	Ash influences the overall power plant efficiency mainly via decreased flame temperature, decreased heat absorption in heat exchangers, and increased deposits of ash as slag on the boiler heating surfaces. A decrease in the overall power plant efficiency is expected to increase impacts on climate change, as more carbon has to be fired per unit of electricity produced, which will increase overall CO ₂ emissions.
Increased life time of the plant	Reduction in ash content reduces fouling and corrosion of power plant components which increases the life time of the plant ^{24,25} . This is expected to decrease the need for steel and metals and thereby reduce impacts on resource (minerals) depletion.
Reduced amount of coal transported between the cleaning site (typically at mine) and the power plant	Chemical cleaning increases the higher heating value (HHV) of the coal, therefore less coal has to be transported from the cleaning site to the power plant per unit of electricity produced ²⁶ . This will reduce fuel consumption for the transport and will thereby decrease the associated emissions of CO ₂ and PM. This is expected to lead to reduced fossil depletion and climate change impacts and respiratory effects on humans.
<i>Expected increase in environmental impacts:</i>	
Need for heat and electricity for cleaning	Increased emissions of CO ₂ and metals from energy production are expected to increase climate change impacts and toxic impacts on human health and ecosystems, depending on the energy source.
Need for NaOH, acids, methanol and water for cleaning	Extraction of raw materials and production of feedstock will lead to increased demand for natural gas and water. This is for instance expected to increase land use impacts and fossil depletion from natural gas extraction.

2. Methods

2.1. Life cycle assessment

The LCA was conducted in accordance with the requirements of the ISO standard²⁷ and the guidelines of the ILCD handbook²⁸. The functional unit is defined as the “output to high voltage grid of 1 MJ of electricity produced from a pulverized coal power plant”. Power generation from chemically cleaned coals using various acid or alkali-acid leaching procedures was compared to power generation from coals cleaned physically using dense medium separation (gravity-based process), and to power generation from raw coals. Power generation from coal cleaned using dense

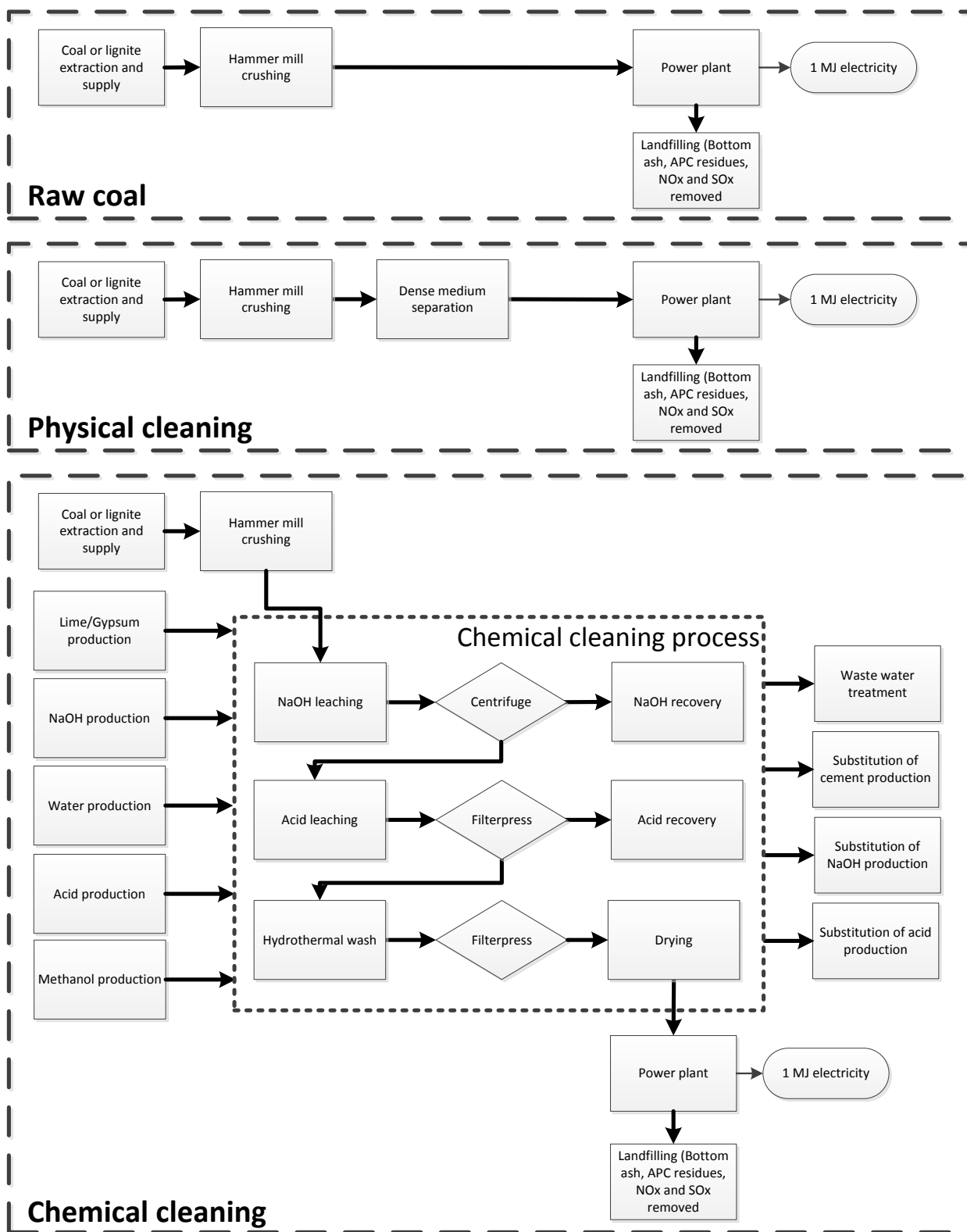
153 medium separation is the most widely used physical cleaning method of coal. Power generation
154 from high-ash raw coal is rare, but is reported to take place in India¹³.

155 Europe was chosen as the primary geographical scope of the assessment. However, as part of a
156 sensitivity check, the comparisons were made using U.S.A. and China as alternative regions to test
157 the validity of our conclusions and whether they change for other geographical regions. U.S.A and
158 China were chosen as these are among the largest coal users worldwide²⁹. The system boundaries
159 include the whole life cycle of the power generation, from the construction of the PC power plant
160 and the extraction and supply of bituminous coal, sub-bituminous coal and lignite, through the
161 cleaning process, to coal firing the plant, slag disposal, and decommissioning of the plant. In
162 chemical cleaning, coal is first crushed using a hammer mill as crusher, and then leached in one step
163 with an acid (acid leaching) or in two steps with first alkali and then acid (alkali-acid leaching) to
164 dissolve and remove inert materials in the ash (see ESI † section S1 for details on chemical
165 cleaning). In physical cleaning, coal is first crushed using a hammer mill and is next washed with
166 water to remove inert impurities that are easily accessible and not embedded in the coal matrix^{30,31}.
167 Raw coal is only crushed with a hammer mill before firing, but no washing procedure is applied.

168 The ILCD guidelines provide methodological guidance according to different decision
169 situations, and the current study is in this context considered a micro-level decision support (type-
170 A) situation. The use of chemical demineralization for cleaning of coal is not expected to have large
171 structural changes on the market (*e.g.* installation of new coal firing capacity or opening new coal
172 mines) compared to current coal practice, at least at the current state of maturity and operation of
173 the chemical cleaning technology. Therefore, the comparison applies an attributional LCA
174 approach, where average data and energy mixes are used. Bituminous and sub-bituminous coals are
175 modelled to represent the European average situation where 78 % of the coal is extracted in Europe,
176 and the rest is extracted in many parts of the World outside Europe. Lignite combusted in Europe is
177 extracted in European countries only, primarily in Germany, Greece and Poland. Transport via
178 shipping from extraction locations outside of Europe is included, as is storing and raw coal
179 pretreatment, *i.e.* washing). A European average electricity grid mix is used for the energy supply to
180 the cleaning process and for the coal handling after it has been transported to Europe. Electricity
181 and heat production are modelled as a European average. Alkali and acids that are produced in
182 Europe are modelled for European conditions. Globally produced and traded commodities such as
183 metals needed for construction are modelled as global production. The PC power plant and the
184 subsequent slag treatment are modelled for European conditions. In cases of processes with

185 recovery of commodities, system expansion was performed, assuming that recovered chemicals
186 (acid and alkali) substitute the production of virgin chemicals, and that ash extracted from coal
187 during chemical cleaning, being a lime product, substitutes virgin lime and gypsum as input for
188 cement production ³². For U.S.A. and China, different processes were used for coal extraction,
189 electricity generation, and PC power plant technology (see Table S1 in ESI † for more information
190 on the geographical scope).

191 The product systems were modeled in the LCA software GaBi, version 4.3 (PE International,
192 Germany). Environmental impact scores were calculated using the ReCiPe (version 1.05)
193 characterization factors, as implemented in GaBi.



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Fig. 1. System boundaries for power generation from raw coal (upper), for coal cleaned using physical cleaning (middle) and for coal cleaned using chemical cleaning with acid or alkali-acid leaching (lower).

2.2. Data collection and model parameters

Data on types of equipment for chemical cleaning are based on the full-scale process patented by Brooks et al.³³, and are combined with data on leaching process conditions, including types and concentration of acids and/or alkali, reaction temperature and duration. This data was retrieved from published studies, available up to September 2014, identified using ISI Web of Knowledge (version 5.7; Thomson Reuters, New York, NY), and were included if the following criteria were met: (i) types of alkali and/or acid are reported, (ii) alkali and/or acid concentrations, cleaning process temperatures and cleaning duration are reported, (iii) initial ash, sulfur, nitrogen, carbon and oxygen contents in the coal are reported, and (iv) final ash contents of the coal are reported. Cited and citing studies that were found to contain relevant data were then consulted to complement the search, and this process was iterated until no new study was found. Studies on cleaning with organic solvent extraction and those with irradiation (such as microwave or ultrasonic) as leaching pretreatment, were excluded. In total, 10 studies (^{14–18,34–38}) were found which met the data requirement criteria. They contain 132 data points for alkali-acid leaching and 107 data points for acid leaching, with 151, 36 and 52 measured data points for bituminous, sub-bituminous and lignite coals, respectively (see section S3 in ESI † for details of the data). Calculated higher heating values (HHV) ranged from 14.9 to 29.5 MJ/kg, depending on the type of coal and the final ash content (Fig. 2). Ash removal efficiencies ranged from 0.05 to 0.97 across all coals, and were the highest for chemical cleaning of high-ash lignites (data not shown).

Model parameters for the processes included within the system boundaries are synthesized in Table 2 and references given. Data for the PC power plant are based on the inventory for a subcritical PC power plant as included in ecoinvent v2.2³⁹ and were modified to reflect the differences between different types of coal with regard to combustion efficiency and emission of pollutants. During combustion, all C, S and N compounds were assumed to be oxidized to CO₂, SO_x and NO_x. Emissions of these gases depend on the installed flue gas cleaning systems, which vary between plants (see Table 2).

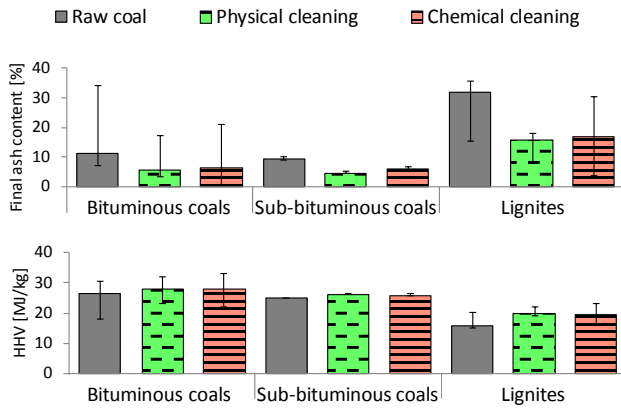


Fig. 2. Final ash content and HHV values of raw and cleaned coals. Error bars indicate the 95 % variability intervals. The HHV was calculated using the formula

$HHV = 0.341 \times C + 1.323 \times H + 0.0685 - 0.0153 \times Ash - 0.1194 \times (O + N)$ which is based on 700 coal samples, where C , H , O , N and Ash are fractions of the respective element and ash contents in the coal (in $\text{kg}_{\text{element}}/\text{kg}_{\text{coal}}$ and $\text{kg}_{\text{ash}}/\text{kg}_{\text{coal}}$, respectively)⁴⁰.

230 **Table 2.** Model parameters and data sources.

Parameter	Average (min - max)	Unit	Note	Source
Coal power plant				
Power plant efficiency at standard ash content (E_0)	0.366 (0.260 – 0.430)	MJ/MJ	The power plant efficiency at standard (6 %) ash content (E_0). Assumed equal to energy efficiency across subcritical, supercritical and ultra-supercritical power plants in EU member states (see Table S3 in ESI †). The average and variability ranges for Europe were also applied for power plants in China and U.S.A. This choice does not influence the main goal of this study <i>i.e.</i> the comparison between coal cleaning technologies, but will to some extent influence the environmental performance of coal burning in general	⁴¹
Overall power plant efficiency (E)	0.362 (0.245 – 0.432)	MJ/MJ	The overall efficiency of the power plant (E) in MJ electricity produced per MJ of thermal input. Calculated as a product of the plant-specific efficiency at standard ash content (E_0) and the coal-specific, ash-dependent efficiency ratio (ER) using the formula $E = E_0 \cdot ER$ (²⁴)	calculated
Efficiency ratio (ER)	0.988 (0.941– 1.005)	(-)	The efficiency ratio takes into account effects of ash content (<i>Ash</i> , in fraction) on the boiler efficiency, auxiliary efficiencies, steam efficiency, turbine efficiency and generator efficiency. It increases with decreasing ash content. Calculated using the formula $ER = 1.005 - 0.05189 \times Ash - 0.52841 \times Ash^2$ (²⁴)	calculated
Flue gas cleaning, NO _x removal efficiency	80 (75 - 85)	%	Range of values measured for NO _x removal using selective catalytic reduction (SCR) process, being one of the most common post-treatment methods.	²²
Flue gas cleaning, SO _x removal efficiency	89 (80 - 98)	%	Range of values measured for SO ₂ removal using flue gas desulfurization (FGD), that utilizes a variety of slurry or sorbent materials to scrub gases	²²
Flue gas cleaning, PM removal efficiency	97 (95 – 99)	%	Range of values for coal power plants, where on average 3 % of initial ash content is emitted to air as particulate matter and the remaining is captured and collected as bottom ash and landfilled	ecoinvent v2.2 ³⁹
Chemical cleaning using acid or alkali-acid leaching procedures				
Centrifuge, electricity use	1.5 (0.03 – 3.21)	MJ/m ³ liquid	The electricity required for removing liquid from the coal slurry. Values measured for high- and low-gravity centrifuges of sizes 74 × 208 and 90 × 225 (cm diameter × 208 cm length), respectively ⁴² .	assumed
Filter press, electricity use	0.88 (0.66 – 1.10)	MJ/m ³ liquid	The electricity required for removing liquid from the coal slurry. Measured for algae recovery ⁴³ and assumed similar in energy used for coal slurry.	assumed
Solid content in slurry after filter press	42.5 (35 – 50)	%	The solid content present in the coal slurry after filter press, measured for batch filter presses used for dewatering wastewater with high solids content ⁴⁴	assumed
Hydrothermal washing heat use	3573	MJ/m ³ slurry	The energy for heat and electricity used for running the hydrothermal washing process, calculated from heat equation for a mixture of water, methanol and coal to heated up to 240 °C, as explained in the ESI † section S5	calculated
Water to methanol ratio for hydrothermal washing	1:1	kg/kg	The ratio in which methanol and water are mixed for the hydrothermal washing. A solvent such as methanol is stated as a requirement by Brooks et al. ³³ and is applied in the base scenario. However, the literature providing the data points used is not reporting whether a solvent is used during washing of the coal, and in many cases only water is applied. We therefore perform a sensitivity test using different ratios of methanol to water	³³
Liquid (water + methanol) to coal ratio for hydrothermal washing	4:1	kg/kg	The default mixing ratio of water and methanol liquid with coal for the hydrothermal washing as used in the base scenario	³³
Coal drying, heat use	8.1	MJ/kg water	The energy required for heating and removing water via drying to obtain clean water-free coal	⁴⁵
Ambient liquid temperature	12	°C	The ambient temperature of the liquid used for coal leaching. This affects the energy requirements for heating the water during demineralization.	assumed
Reagents recovery efficiency	85	%	Alkali and the acids are recovered after the leaching process using lime and gypsum, respectively, The efficiency of the recovery process for recovery of alkali and acid is based on one source only, hence no range is provided. The recovery reactions are as follows:	⁴⁸

			Alkali recovery: $Na_2SiO_3 + Ca(OH)_2 \rightarrow Ca_2SiO_3 + 2NaOH$ Acid recovery: $H_2SiO_3 + CaSO_4 \rightarrow CaSiO_3 + H_2SO_4$ <small>32,46,47</small>	
Reagents recovery electricity use	0.005	MJ/kg	The electricity use for recovery of alkali and acids	⁴⁸
Acid or alkali concentration				
HCl	11.6 (5.0 – 30.0)	%	Aqueous concentration of HCl in the acid leaching step	various sources (146 data points, see Table S2 in ESI †)
HNO ₃	19.0 (5.0 – 30.0)	%	Aqueous concentration of HNO ₃ in the acid leaching step	various sources (26 data points, see Table S2 in ESI †)
H ₂ SO ₄	15.5 (5.0 – 30.0)	%	Aqueous concentration of H ₂ SO ₄ in the acid leaching step	various sources (43 data points, see Table S2 in ESI †)
HF	9.5 (1.3 – 25.5)	%	Aqueous concentration of HF in the acid leaching step	various sources (14 data points, see Table S2 in ESI †)
H ₂ O ₂	12.5 (2.5 – 30.0)	%	Aqueous concentration of H ₂ O ₂ in the acid leaching step	various sources (10 data points, see Table S2 in ESI †)
NaOH	28.5 (2.0 – 98.0)	%	Aqueous concentration of NaOH in the alkali leaching step	various sources (132 data points, see Table S2 in ESI †)
Temperature of the acid leaching step	84.5 (25.0 – 100.0)	°C	The temperature during the acid leaching	various sources (239 data points, see Table S2 in ESI †)
Duration of acid leaching step	2.4 (0.2 – 8.0)	hours	The duration of the acid leaching	various sources (239 data points, see Table S2 in ESI †)
Temperature of alkali leaching step	146.4 (85.0 – 210.0)	°C	The temperature during the alkali leaching step	various sources (132 data points, see Table S2 in ESI †)
Duration of alkali leaching step	1.7 (0.5 – 24.0)	hours	The duration of the alkali leaching step	various sources (132 data points, see Table S2 in ESI †)
Sulfur reduction efficiency due to chemical cleaning	20 (9 – 48)	%	Reduction of sulfur content in the coal as a function of alkali-acid leaching predicted from NaOH concentration, acid concentration, time of alkali leaching and time of acid model using a model developed based on literature data (see ESI † section S6). The model has been validated against external data, and its predictive power corresponds to a predictive squared correlation coefficient for external validation (Q^2) equal to 0.8.	calculated
Nitrogen reduction efficiency due to chemical cleaning	0	%	No nitrogen reduction considered, as literature on nitrogen content from chemical demineralization is inconclusive. A number of sources state that nitrogen content increases with alkali-acid leaching ^{19,49,50} . This has only been shown where nitric acid is used, in these cases the nitrogen containing acid may react to form nitrogen-bearing species within the coal ⁵⁰ .	assumed
Physical cleaning using dense medium separation (gravity-based method)				
Ash removal efficiency	50 (16 – 68)	%	The ash removal efficiency from the coal after physical cleaning using dense medium separation measured for various dense medium separation procedures.	⁵¹
Sulfur removal efficiency	48 (10 – 84)	%	The sulfur removal efficiency from the coal after physical cleaning measured for various dense medium separation procedures.	⁵¹
Electricity use	0.027 (0.022 – 0.032)	MJ/kg coal treated	The electricity use for physical cleaning of coal at mine measured for a dense medium separation procedures process.	⁵²
Transportation				
From coal storage to power plant	100	km	By lorry	assumed
From coal storage to demineralization plant	10	km	By lorry	assumed
From demineralization plant to power plant	100	km	By lorry	assumed

2.3. Sensitivity analysis

Sensitivity of impact scores to uncertain or variable model parameters was done by calculating normalized sensitivity coefficients (S_{coef}), according to Eq. 1^{53,54}:

$$S_{coef} = \frac{\Delta IS}{IS_0} \bigg/ \frac{\Delta a_k}{a_{k,0}} \quad (1)$$

where $a_{k,0}$ is the input parameter value, IS_0 is the impact score calculated for the $a_{k,0}$, Δa_k is the difference between the default input parameter and the perturbed input parameter, ΔIS is the difference between IS_0 and the impact score calculated for the perturbed parameter value. All input parameters were perturbed by 25%. All parameters in Table 2 were included in the sensitivity analysis. Among all 239 chemical cleaning combinations, the S_{coef} was calculated for a total of 16 combinations, selected to represent a range of chemical cleaning process conditions, (*i.e.* the highest and the lowest values of: NaOH concentration, acid concentration, time of the alkali leaching step, temperature of the alkali leaching step, time of the acid leaching step, and temperature of the acid leaching step), a range of initial ash contents (*i.e.* the highest and lowest initial ash content) and a range of ash removal efficiencies (*i.e.* the highest and the lowest ash removal efficiency). A parameter is considered important if average $|S_{coef}| \geq 0.3$, or if the largest $|S_{coef}| \geq 0.5$, corresponding to a medium and large sensitivity, respectively⁵⁵.

2.4. Uncertainty and variability analysis

Parameter uncertainties stem from the lack of knowledge about the actual value of a parameter⁵⁶, *e.g.* electricity use of a centrifuge. By contrast, variability is the inherent variance that will exist between similar processes depending on technological level and spatial location, *e.g.* power plant efficiency at standard ash content⁵⁶. Here, parameter uncertainty was assessed together with variability by means of a Monte Carlo analysis, using parameters which were found important in the sensitivity analysis (Table 3). They were assigned standard deviations based on the min-max ranges in Table 2, accounting for their uncertainty and variability. Normal distributions were assumed. Differences in impact scores between the compared systems were considered significant if the calculated 95% probability ranges of the impact scores from 1000 iterations did not overlap⁵⁷.

Table 3. Uncertain or variable parameters included in the Monte Carlo simulation and the associated relative standard deviation.

Uncertain or variable parameter	Average (relative standard deviation)	Note
Efficiency of NO _x removal	0.8 (6%)	Based on literature ²²
Efficiency of SO _x removal	0.89 (10%)	Based on literature ²²
Power plant efficiency	0.366 (12%)	Mean and standard deviation are based on power plant overall efficiencies of European countries ⁴¹
Centrifuge electricity use	1.5 (30%)	Based on literature ⁴²
Filter press electricity use	0.88 (25%)	Based on ⁴³ , because variability ranges were not available, a relative standard deviation of 25 % is assumed to account for large variations
Sodium hydroxide recovery efficiency	0.85 (25%)	Based on ⁴⁸ , because variability ranges were not available, a relative standard deviation of 25 % is assumed to account for large variations, varying between a recovery efficiency of 64 – 100 %
Acid recovery efficiency	0.85 (25%)	Based on ⁴⁸ , because variability ranges were not available, a relative standard deviation of 25 % is assumed to account for large variations, varying between a recovery efficiency of 64 – 100 %
Physical ash removal efficiency	0.50 (33%)	The fraction of ash left in coal after mechanical treatment at mine, using crushing and gravity separation ⁵¹
Mechanical sulfur removal efficiency	0.52 (26%)	The fraction of sulfur left in coal after mechanical treatment at mine, using crushing and gravity separation ⁵¹

3. Results and discussion

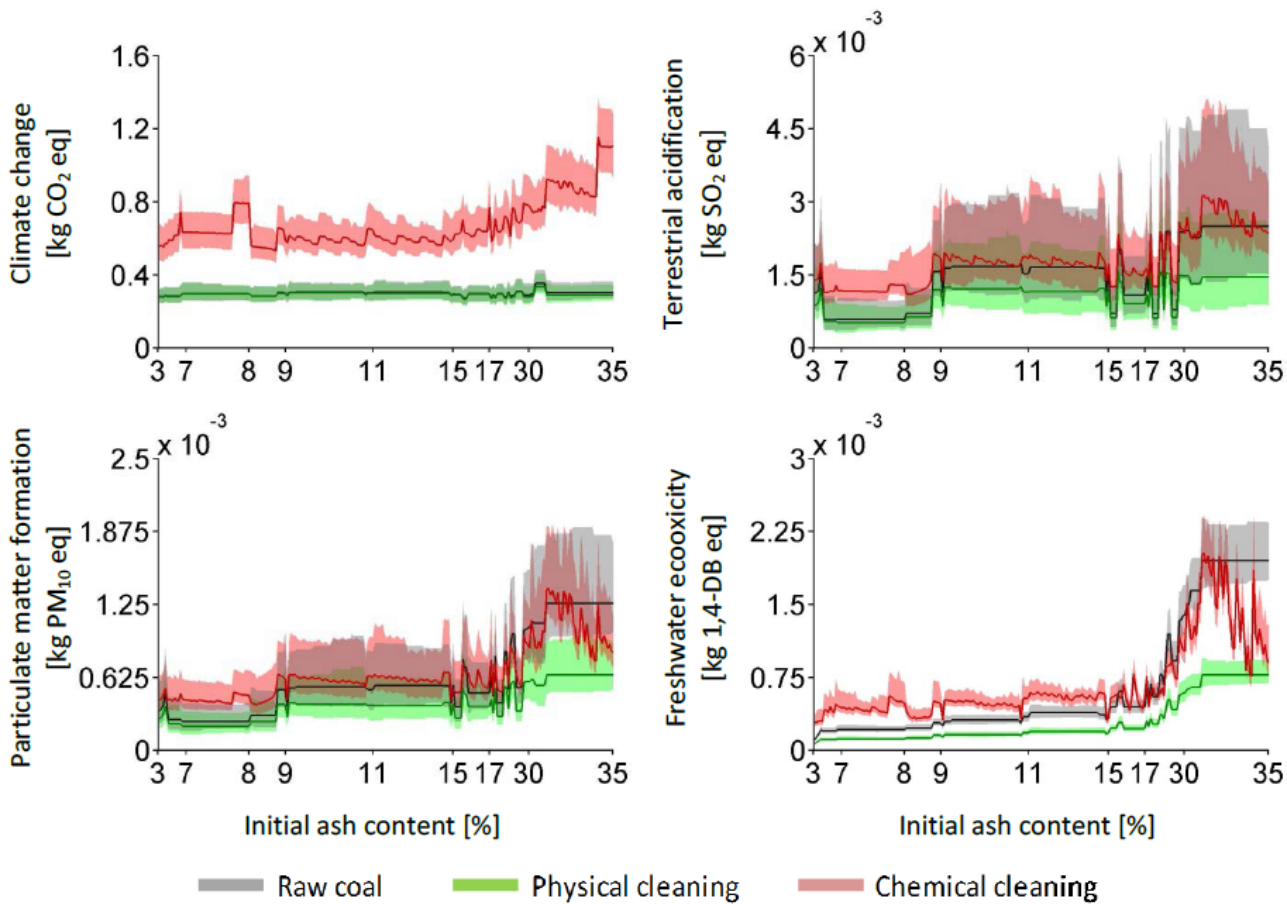
Below, we present results for four selected impact categories, which represent typical impact profiles observed for all 18 impact categories (results for all impact categories are presented in Fig. S2 in ESI †). Next, the potential of the chemical cleaning technology as a coal beneficiation method, and the broader applicability of our findings, are discussed.

3.1. Does coal cleaning bring environmental benefits?

Fig. 3 shows that, irrespective of the initial ash content, coals cleaned chemically perform significantly worse in an overall life cycle perspective than raw coals and coals cleaned physically for the impact category climate change. For acidification impacts on terrestrial ecosystems and impacts from PM on human health, impact scores overlap, whereas for freshwater ecotoxicity (toxic impacts on freshwater ecosystems), the environmental performance of the chemically cleaned coal depends on the initial ash content of the coal: for low-ash coals chemical cleaning performs worse as compared to raw coal, and the opposite is observed for high-ash coals. Further, freshwater ecotoxicity impact scores for coals cleaned chemically peak at 30% of initial ash content.

282 The increase in impact scores with initial ash content up to 30% for freshwater ecotoxicity (but
 283 also for freshwater eutrophication, PM formation and other toxicity-related impact categories, see
 284 Fig. S2 in ESI †) is primarily caused by emissions of particles and leaching of heavy metals and
 285 phosphate from landfilling of residual (bottom) ash. These emissions are naturally higher for high-
 286 ash coals. Thus, there is some potential for avoiding environmental problems caused by firing coals
 287 if the ash is separated from the coal before the firing. Indeed, the decrease in impact scores for
 288 chemically cleaned high-ash coals (containing > 30% ash in raw coal) is due to a very efficient
 289 cleaning of lignites, for which up to 90% ash removal efficiency can be reached using acid or alkali-
 290 acid leaching procedures¹⁷. In contrast, the ash removal efficiency for physical cleaning generally
 291 does not depend on the coal type, which explains why environmental performance of high-ash
 292 lignites cleaned chemically improves relative to these coals cleaned physically.

293
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295
 296 **Fig. 3.** Impact scores and the associated 95% confidence intervals per functional unit (“output to
 297 high voltage grid of 1 MJ of electricity produced from a pulverized coal power plant”) as a function

of ash content in the raw coal, shown for each of the four selected impact categories. The results are shown for the European scenario.

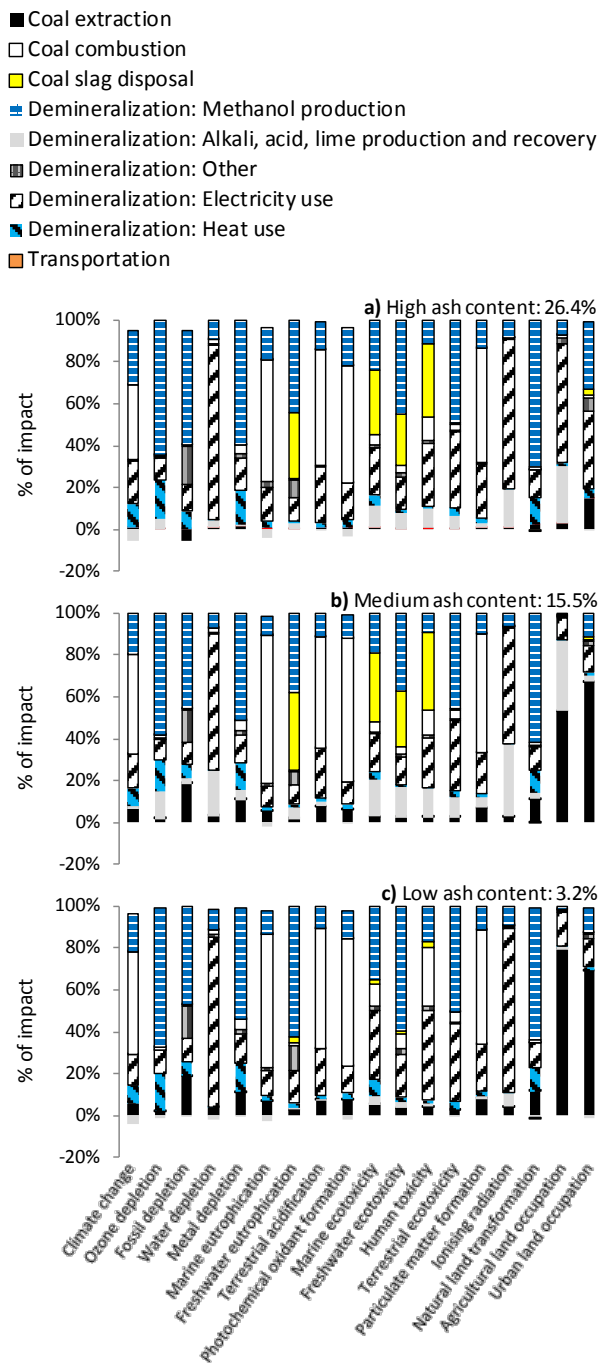


Fig. 4. Contribution of life cycle processes to total impact scores for coal firing using chemical cleaning (alkali-acid leaching) of coals with different initial ash contents. The results are shown for the European scenario.

3.2. Can chemical cleaning become an environmentally sound approach for coal beneficiation?

Despite the potential benefits of chemical cleaning of high-ash coals for some impact categories, our results show that burden shifting might occur if only environmental problems directly caused by ash are considered. Indeed, impact scores for other relevant impact categories including climate change and depletion of resources (land, water, minerals and fossils) are increased when ash is removed before firing (see Fig. 3 and Fig. S2 in ESI †). Such trends are caused by non-ash-related emissions, which have large contributions in other environmental impacts and stem from the cleaning process. For example, while the reduction in PM emissions from the firing of the chemically-cleaned lignites outweigh the increase in PM emissions from the cleaning processes, thus resulting in an overall decrease of impact scores for the PM formation impact category, the opposite trend is observed for climate change and other impact categories (see Fig. 3). Improvements in the environmental performance of chemical cleaning should therefore focus on optimization of cleaning procedures in order to minimize all relevant environmental impacts.

To identify improvement potentials for the chemical cleaning technology a process contribution analysis was conducted, *i.e.* identifying the processes with the largest environmental burden. Fig. 4 shows that the largest contributors to environmental impacts are (i) the electricity consumption for centrifugation in the alkali step of the leaching procedure, and (ii) the production of methanol for the hydrothermal washing step of the cleaning procedure. The contributions of these two processes were above 50% of the total impact score for many impact categories. The sensitivity analysis further confirmed that these processes had the largest influence on impact scores (see Table S6 in ESI †). Hence, there is a potential for improving the environmental performance of the chemical cleaning technology if the electricity demand for chemical demineralization and the methanol use for the hydrothermal washing step are optimized.

In our study of chemical cleaning we had to rely on data from the pilot scale process combined with parameters retrieved from laboratory measurements and estimations based on physical relationships (e.g. energy required for heating a liquid solution). However, commercial scale processes are often seen to have smaller environmental impacts due to the use of more efficient use of processes and feedstock⁵⁸ compared to laboratory scale where efficiency is less important and focus is on developing the cleaning process and achieving useful results. Thus, it cannot be ruled out that the overall demand for chemicals and energy will be smaller when chemical cleaning is performed at commercial scale. Because chemical cleaning is still a relatively new and immature

338 technology some improvement with regard to use of energy and feedstock as a result of upscaling
339 can be expected. Yet, our analysis shows that a reduction in total electricity and heat use in
340 chemical cleaning by a factor of 5 to 10, depending on the impact category would make chemical
341 cleaning perform comparably to physical cleaning; this is illustrated in Fig. 5-1 for the climate
342 change impact category. Achieving such reduction poses a strong challenge for the technology and
343 seems unlikely. For petrochemical production processes, which share some similarities with
344 chemical cleaning (e.g. centrifuge, filtering and heating), the energy consumption can be reduced by
345 16 % - 20 % by adopting best available technologies (BAT) ^{59,60}. The heat demand from upstream
346 sources can be reduced by utilizing low grade waste heat (energy pinching), which is estimated to
347 increase energy efficiency by circa a factor 1.2 ⁶¹⁻⁶⁴, while electricity efficiency may be increased
348 by use of kinetic energy recovery systems or improving the efficiency of electricity using systems,
349 this can yield electricity savings between 5 % - 40 % ^{64,65}.

350 Methanol is produced from natural gas, through synthesis gas, which is then converted to
351 methanol using metal oxide catalysts ^{39,66}. Because methanol is a key intermediary product and is
352 vital for the production of many globally-used commodities (e.g. formaldehyde and solvents),
353 finding cleaner alternatives for methanol appears unlikely. However, the use of a solvent such as
354 methanol during the hydrothermal washing step is not absolutely necessary; Mukherjee ⁶⁷ already
355 showed that alkali-acid cleaning combined with washing in a methanol solution did not show
356 improved cleaning efficiencies compared to similar alkali-acid leaching studies where the same coal
357 was washed with pure water ^{18,34,38}. We therefore tested if excluding methanol from the cleaning
358 procedure improves environmental performance of chemical cleaning. Figure 5-2 shows that
359 although methanol contributes substantially to a number of impact categories, the influence of
360 excluding it from the hydrothermal washing step on impact scores is modest. This is because
361 chemical cleaning generally perform worse than physical cleaning for the impact categories where
362 methanol is found to be an important contributor, and whether methanol is included makes little
363 difference on the comparison (see ESI Fig. S2). Thus, while phasing out methanol and replacing it
364 with water is overall an important improvement of the cleaning procedure, it is not sufficient for
365 chemical cleaning to perform better as compared to physical cleaning

366 Developers of leaching procedures typically focus on optimizing reaction conditions and
367 selection of appropriate acids, which in this study are seen to have relatively small contribution to
368 total environmental impacts and are not seen to influence impact scores significantly (S_{coef} typically
369 below 0.1, see Section S9 in ESI †). Thus, from the environmental performance perspective, higher

370 concentrations of acids and alkali and higher leaching temperatures and durations can be justified if
371 they allow for increasing ash removal efficiencies and thereby increasing the environmental benefits
372 associated with firing cleaner coal. This is observed for lignites, of which many have their initial
373 ash content equal to 35%: more severe cleaning conditions with regard to temperatures and
374 concentrations of chemicals do not translate into higher environmental impacts.

375 In summary, chemical cleaning is not likely to become an environmentally sound approach for
376 coal beneficiation, unless developers of chemical cleaning procedures (i) use water instead of
377 methanol in the washing step, (ii) focus on achieving very high ash removal efficiencies by
378 optimizing the use of alkali and acids, and (iii) find more attractive, environmentally speaking,
379 ways of separating liquid from coal slurry after each washing steps.

380

381 **3.3. Applicability of the findings for other geographic locations**

382 Our conclusions about the overall poor performance of chemical cleaning are not expected to
383 change when assuming coal power production in U.S.A. or China. Even though impact scores for
384 these two countries are higher for many impact categories as compared to Europe, chemical
385 cleaning is generally seen as the worst alternative in terms of environmental impacts. However, for
386 terrestrial acidification impacts (see Fig. 5), PM formation and marine eutrophication in the Chinese
387 scenario, impact scores for chemical cleaning are lower than those for the raw coal (data not
388 shown). Chinese plants in our model have lower SO₂ cleaning efficiencies and PM removal
389 efficiencies. Hence chemical cleaning becomes environmentally beneficial as it allows removing a
390 part of sulfur and ash from the coal that otherwise would have been emitted as SO₂ and PM. This
391 could suggest that chemical cleaning could be an environmentally sound technology for ash
392 removal in regions where pollutant emission regulations are not as stringent as in Europe. However
393 environmental burden shifting might still occur as for other impact categories, just as for climate
394 change, chemical cleaning performs worse than physical cleaning or firing of raw coal.

395 The overall power plant efficiency was by far the most influential parameter (with S_{coef} equal to
396 -0.8 for all impact categories), hence, an increase in the overall power plant efficiency will reduce
397 environmental burdens associated with power generation. However, simply shifting to a power
398 plant with a higher overall efficiency will not make chemical cleaning an environmentally sound
399 coal beneficiation options, unless large reductions in combustion efficiencies due to high ash
400 contents are observed for highly-efficient plants. Such situations, however, do not apply to PC
401 power plants. Yet, a reduction of ash content below 0.1% allows coal firing in an integrated

gasification combined cycle. Such application supports our findings that chemical cleaning technology may be environmentally beneficial when used to clean high ash coals with very high removal efficiencies (see Section 3.2).

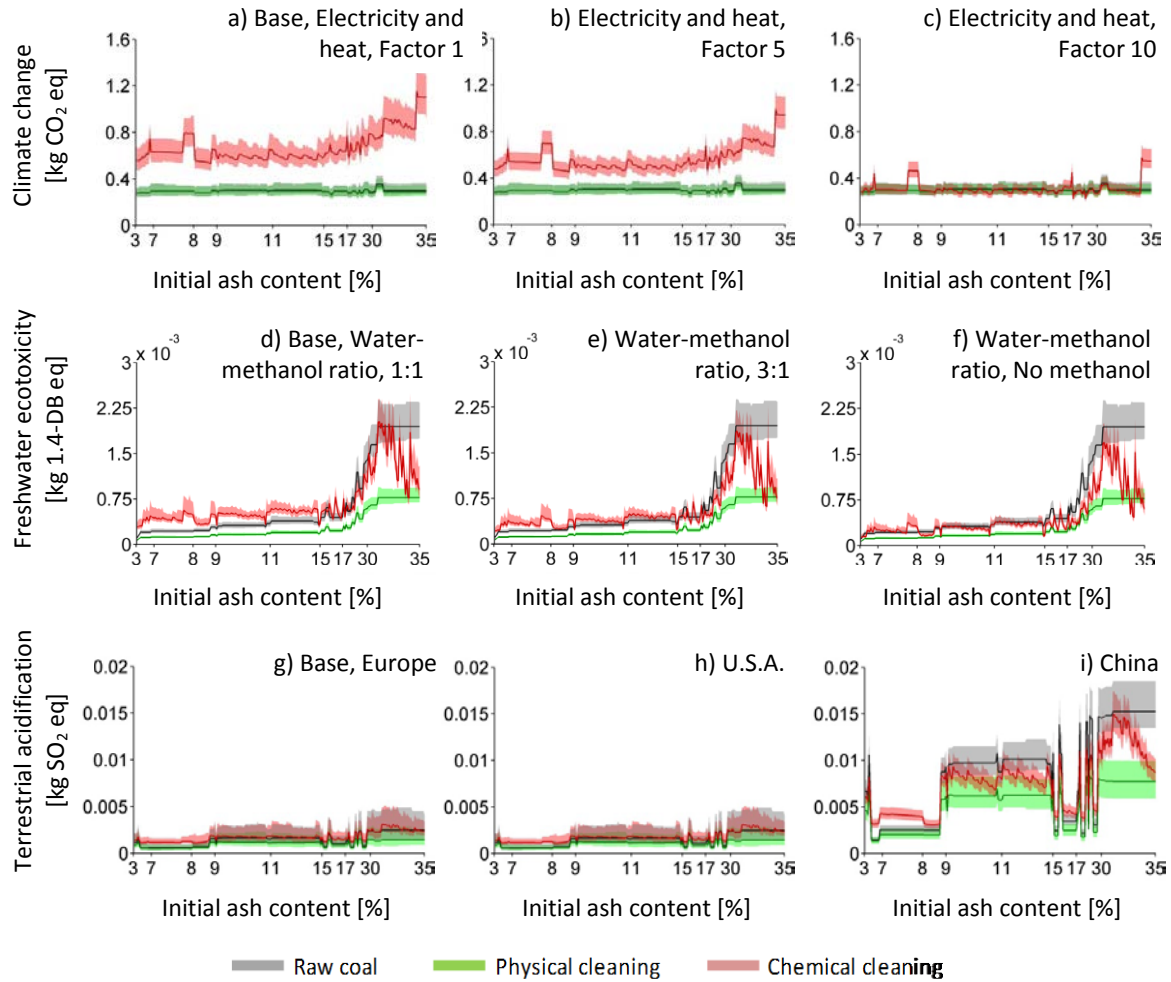


Fig. 5. Impact scores for climate change in the European scenario are shown for **a)** the base scenario, **b)** for the scenario where the electricity and heat use for chemical cleaning was reduced by a factor 5 and **c)** for the scenario where the electricity and heat use was reduced by a factor 10. Underneath, impact scores for freshwater ecotoxicity in the European scenario are shown in **d)** for the base scenario using 1:1 water to methanol ratio, in **e)** using 3:1 water to methanol ratio and in **f)** where methanol is excluded and only water is used during the hydrothermal wash. Finally, impact scores for terrestrial acidification are shown for three different geographical scopes **g)** the base scenario *i.e.* Europe, **h)** U.S.A. and **i)** China.

4. Conclusions and recommendations for policy makers

The increased use of low rank coals for power generation has resulted in the implementation of stricter legislation on ash and sulphur contents in the coal, primarily to reduce air pollution and transport costs. For instance, as part of the “Interim Measures on the Management of Commercial Coal Quality“, China has imposed restrictions on ash and sulphur contents in lignites for commercial use which must stay below 30% and 1.5% respectively to reduce emissions from coal burning⁶⁸. India has banned coal with ash content above 34 % in order to reduce impacts and costs from the often long transports inside the country ¹⁰. Increased research and development of chemical coal cleaning has been recommended in a report to the U.S. Secretary of Energy by the National Coal Council, NCC ⁶⁹(which serves as an advisory council to the Secretary of Energy and NCC members comes from both industry and academia). This new legislation is expected to increase demand for chemical cleaning technologies in the future, particularly for high-ash coals with ash and sulphur being strongly embedded in the coal matrix, for which physical cleaning is not sufficient method for cleaning. Yet, as we showed that chemical cleaning of coals for firing in PC power plants in general is not an environmentally sound option for coal beneficiation, we recommend policy makers that coal cleaning using acid or alkali-acid leaching procedures should not be considered for direct implementation as a coal beneficiation technology. We note, however, that chemical cleaning is a relatively immature technology and, as showed, its environmental performance of chemical cleaning is expected to improve when upscaling from the laboratory scale to the commercial scale is done. These improvements can be achieved by phasing out methanol in the washing step, increasing higher ash removal efficiencies by optimizing the use of alkali and acids, and finding more attractive, environmentally speaking, ways of separating liquid from coal slurry after each washing step. When these measures are undertaken, chemical cleaning might become environmentally attractive technology that can complement physical cleaning methods for ash removal for high-ash, difficult to treat coals. We stress that life cycle based approaches, such as LCA must be used to determine for which coals and cleaning procedures, chemical cleaning can be considered as potential beneficiation technology to avoid environmental burden shifting which occurs when environmental benefits of firing cleaned coal do not outweigh environmental burden of the cleaning. Finally, for high-ash coals where ash is relatively easy to remove and for coals with low ash content, we recommend policy makers to focus on physical methods of cleaning. They clearly perform better in a life cycle perspective as compared to chemical cleaning or combustion of

raw coal and it is not likely that they will be able to compete with physical cleaning in terms of environmental performance even when eco-design measures are undertaken.

5. Notes and References

† Electronic Supplementary Information (ESI) available: Supplementary Methods (Figure S1; Tables S1-S4) and Supplementary Results and Discussions (Figure S2; Tables S5-S7).

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